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# New prospects in synthesis and study of neutron rich heavy nuclei

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**Abstract.** The present limits of the upper part of the nuclear map are rather close to the beta stability line while the unexplored area of heavy neutron rich nuclides (also those located along the neutron closed shell $N = 126$ to the right hand side of the stability line) is extremely important for nuclear astrophysics investigations and, in particular, for the understanding of the r-process of astrophysical nucleogenesis. For elements with $Z > 100$ only neutron deficient isotopes (located to the left of the stability line) have been synthesized so far. The "north-east" area of the nuclear map can be reached neither in fusion–fission reactions nor in fragmentation processes widely used nowadays for the production of new nuclei. Multi-nucleon transfer processes in near barrier collisions of heavy ions seem to be the only reaction mechanism allowing us to produce and explore neutron rich heavy nuclei including those located at the superheavy island of stability. Neutron capture process can be also considered as an alternative method for the production of long-lived neutron rich superheavy nuclei. Strong neutron fluxes might be provided by nuclear reactors and nuclear explosions in laboratory frame and by supernova explosions in nature.

## 1 Motivation

Due to the bending of the stability line forward to neutron axis, in fusion reactions of stable nuclei one may produce only proton rich isotopes of heavy elements. For example, in fusion of rather neutron rich $^{18}$O and $^{186}$W isotopes we obtain the neutron deficient excited compound nucleus $^{204}$Pb, which after evaporation of several neutrons shifts even more to the proton rich side. That is the main reason for the impossibility to reach the center of the "island of stability" ($Z \sim 110 \div 120$ and $N \sim 184$) in the superheavy (SH) mass region in fusion reactions with stable projectiles. Note that for elements with $Z > 100$ only neutron deficient isotopes (located to the left of the stability line) have been synthesized so far. Because of that we also have almost no information about neutron rich isotopes of heavy elements located in the whole "north-east" part of the nuclear map: for example, there are 19 known neutron rich isotopes of cesium ($Z = 55$) and only 4 of platinum ($Z = 78$).

At the same time this unexplored area of heavy neutron rich nuclei is extremely important for nuclear astrophysics investigations and, in particular, for the understanding of the r-process of astrophysical nucleogenesis (a sequence of neutron-capture and $\beta$-decay processes). The origin of heavy elements from iron to uranium remains one of the great unanswered questions of modern physics and it is likely to remain a hot research topic for the years to come. The r-process path is located (and probably interrupted by fission) just in the region of unknown heavy nuclei with a large neutron excess (see Fig. 1).

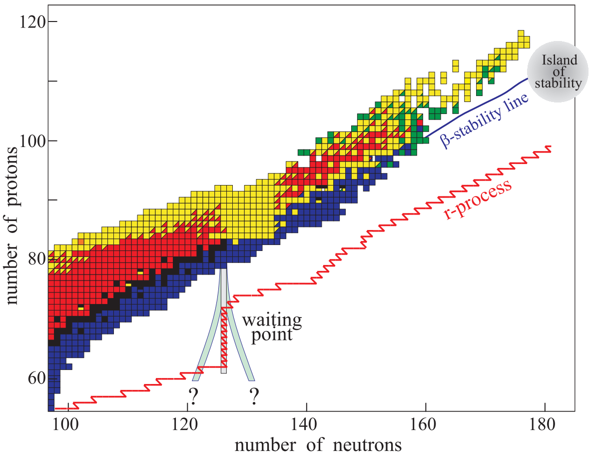

**Fig. 1.** Top part of the nuclear map. The r-process path and the island of stability are shown schematically.

The neutron shell $N = 126$ (and $Z \sim 70$) is the last "waiting point" on this path. The half-lives and other characteristics of these nuclei are extremely important for the r-process scenario of the nucleosynthesis. Study of the structural properties of nuclei along the neutron shell $N = 126$ could also contribute to the present discussion of the quenching of shell gaps in nuclei with large neutron excess. The isotopes with extreme neutron-to-proton ratios in the mass region $A = 80 \div 140$ are successfully produced, separated and studied in fission processes of actinide nuclei, whereas the neutron rich nuclei with $Z > 60$ cannot be formed nei-

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ther in fission nor in fusion reactions. This area of the nuclear map remains blank for many years.

In the history of the synthesis of SH nuclei two significant pages have been overturned within last twenty years. In the "cold" fusion reactions based on the closed shell target nuclei, lead and bismuth, proton rich SH elements up to $Z = 113$ have been produced [1,2]. The "world record" of 0.03 pb in the production cross section of the 113 element has been obtained here within more than half-year irradiation of $^{209}$Bi target with $^{70}$Zn beam [2]. Further advance in this direction seems to be very difficult.

A ten years epoch of $^{48}$Ca irradiation of actinide targets for the synthesis of SH elements is also over. The heaviest available target of Californium ($Z = 98$) has been used to produce the element 118 [3]. Note that the predicted cross sections and excitation functions for all the $^{48}$Ca induced fusion reactions [4] have been fully confirmed by experiments performed in Dubna and later in Berkeley and GSI. To get SH elements with $Z > 118$ in fusion reactions, one should proceed to heavier than $^{48}$Ca projectiles. The strong dependence of the calculated evaporation residue (EvR) cross sections for the production of SH elements on the mass asymmetry in the entrance channel makes the nearest to $^{48}$Ca projectile, $^{50}$Ti, most promising for further synthesis of SH nuclei. The calculated excitation functions for the synthesis of SH elements 119 and 120 in the fusion reactions of $^{50}$Ti with $^{249}$Bk and $^{249}$Cf targets reach maximal values of about 0.05 pb in the 3n and 4n evaporation channels [5].

The yield of superheavy nuclei (number of events per day) depends not only on the cross section but also on the beam intensity, target thickness and so on. In this connection the other projectile–target combinations should be also considered. Most neutron-rich isotopes of element 120 may be synthesized in the $^{54}$Cr+$^{248}$Cm fusion reaction leading to SH nucleus $^{302}$120 with neutron number near to the predicted closed shell $N = 184$. The estimated EvR cross sections for this reaction [5] are quite comparable with the Ti-induced fusion reaction (still there is advantage factor 2 or 3 for $^{50}$Ti+$^{249}$Cf).

As mentioned above, in all these fusion reactions only *proton rich* SH nuclei with a short half-life can be produced located far from the "island of stability" (see Fig. 1). The half-lives of the isotopes of 120 element synthesized in the titanium induced fusion reaction are already very close to the critical value of one microsecond needed to pass through the separator up to the focal plane detector. The next elements ($Z > 120$) being synthesized in such a way might be beyond this natural limit for their detection. Thus, future studies of SH elements are connected with the production of neutron enriched longer living isotopes of SH nuclei.

There are three possibilities for the production of such nuclei. These are the multi-nucleon transfer reactions, fusion reactions with extremely neutron rich radioactive nuclei [5] and rapid neutron capture process. Today the two last methods look unrealizable because of low intensity of radioactive beams and low neutron fluxes in existing nuclear reactors. However, the specifications of the next generation pulsed reactors (needed to bypass the Fermium gap and the gap of short-living nuclei in the region of Z=106÷108 and A∼270 in the neutron capture processes) are of great interest and might be predicted (see below).

A possibility for the production of new heavy neutron rich nuclei in low-energy multi-nucleon transfer reactions is discussed currently in several laboratories (see, for example, [6,7]). Unfortunately, available experimental setups hardly may be used for this purpose and new (rather expensive) equipment has to be designed and installed to discover and examine these nuclei. In this connection, realistic predictions of the corresponding cross sections for different projectile–target combinations as well as detailed calculations of the charge, mass, energy and angular distributions of transfer reaction fragments made in [8] could be quite useful.

## 2 Neutron rich nuclei along the closed neutron shell N=126

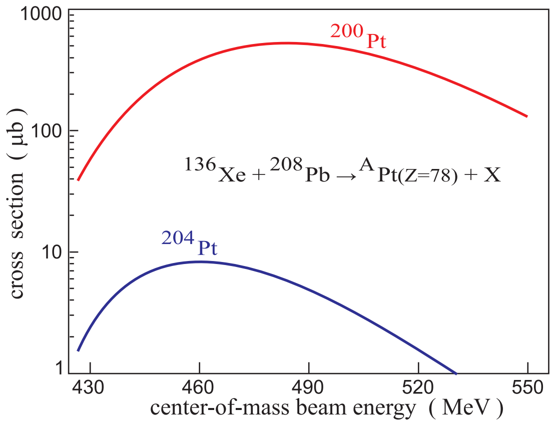

**Fig. 2.** Excitation functions for production of platinum isotopes in collisions of $^{136}$Xe with $^{208}$Pb.

Recently we proposed to take advantage of shell effects for the production of neutron rich nuclei located along the neutron closed shell $N = 126$ ("southward" of doubly magic nucleus $^{208}$Pb) in multi-nucleon transfer processes at low-energy collisions of $^{136}$Xe with $^{208}$Pb target [9]. We found a significant gain in the formation cross sections, which comes from the fact that the reaction $Q$-values remain here close to zero up to 4 transferred protons due to well bound complementary light fragments (having closed neutron shell $N = 82$) formed in these reactions. The low-energy multi-nucleon transfer reactions look more preferable for the production of new heavy neutron rich nuclei as compared to relativistic proton removal processes. However experiments of such kind (prepared now in several laboratories) are rather complicated. At low energies it is more difficult to separate the synthesized new heavy nuclei ($N \sim 126$, $Z > 70$) produced in these reactions. The corresponding excitation functions, energy and angular distributions of heavy reaction products for different projectile–target combinations have been calculated in [8].

The total yield of primary reaction fragments in transfer reactions sharply increases with increasing collision energy. However the excitation energy of these fragments also increases leading to evaporation of more neutrons. Our calculations demonstrated that the excitation functions for the production of neutron rich nuclei are rather wide and have maxima at energies slightly above the Coulomb barrier in the entrance channel. In Fig. 2 the angular integrated yields of survived platinum isotopes (A=200 and 204) are shown for collisions of $^{136}$Xe with $^{208}$Pb target. As can be seen, the production cross section of the neutron rich isotope is maximal at beam energy which exceeds the Coulomb barrier by $20 \div 30$ MeV. The yield of this isotope, $^{204}$Pt, depends strongly on a projectile–target combination. At fixed $^{208}$Pb target it sharply increases with increasing mass number and neutron number of projectile [8].

## 3 Production of transfermium neutron rich nuclei

### 3.1 Multi-nucleon transfer reactions

The multi-nucleon transfer reactions can be used also for the production of new neutron rich isotopes in the SH mass area. Additional enhancement of the corresponding cross sections at low collision energies may originates here due to shell effect. We called it "inverse quasi-fission" process [10]. In this process one of the heavy colliding partners, say $^{238}$U, transforms to lighter doubly magic nucleus $^{208}$Pb while the other one, say $^{248}$Cm, transform to the complementary superheavy nucleus. The role of these shell effects in damped collisions of heavy nuclei is still not absolutely clear and was not carefully studied experimentally. However very optimistic experimental results were obtained recently [11] confirming such effects in the $^{160}$Gd + $^{186}$W reaction for which the similar "inverse quasi-fission" process ($^{160}$Gd→$^{138}$Ba while $^{186}$W→$^{208}$Pb) has been predicted earlier [12].

In multi-nucleon transfer reactions the yields of SH elements with masses heavier than masses of colliding nuclei strongly depend on the reaction combination. The cross sections for the production of *neutron rich* transfermium isotopes in reactions with $^{248}$Cm target change sharply if one changes from medium mass (even neutron rich) projectiles to the uranium beam. In Fig. 3 the mass distributions of heavy primary reaction fragments are shown for near barrier collisions of $^{238}$U, $^{136}$Xe and $^{48}$Ca with curium target. The "lead shoulder" manifests itself in all these reactions. For $^{136}$Xe+$^{248}$Cm and $^{48}$Ca+$^{248}$Cm collisions it corresponds to the usual (symmetrizing) quasi-fission process in which nucleons are transferred mainly from the heavy target (here $^{248}$Cm) to the lighter projectile. Contrary to this ordinary quasi-fission phenomena, for the $^{238}$U + $^{248}$Cm collisions we may expect an inverse process in which nucleons are predominantly transferred from the lighter partner to heavy one (U transforms to Pb and Cm to 106). In this case, besides the lead shoulder in the mass and charge distributions of the reaction fragments, there is also a pronounced shoulder in the region of SH nuclei.

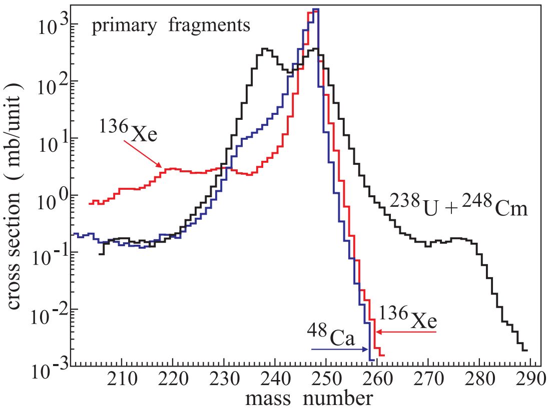

**Fig. 3.** Mass distributions of heavy primary reaction fragments formed in collisions of $^{238}$U, $^{136}$Xe and $^{48}$Ca with $^{248}$Cm at $E_{\mathrm{c.m.}}$=750, 500 and 220 MeV, correspondingly.

### 3.2 Neutron capture processes

The neutron capture process is an alternative (oldest and natural) method for the production of new heavy elements. Strong neutron fluxes might be provided by nuclear reactors and nuclear explosions under laboratory conditions and by supernova explosions in nature. However the "Fermium gap", consisting of the short lived Fermium isotopes $^{258-260}$Fm located at the beta stability line while have very short half-lives for spontaneous fission, impedes formation of nuclei with Z>100 by the weak neutron fluxes realized in existing nuclear reactors. In nuclear and supernova explosions (fast neutron capture) this gap may be bypassed, if the total neutron fluence is high enough. Theoretical models predict also another region of short lived nuclei located at Z=106÷108 and A∼270.

Recently the possibility of synthesizing heavy neutron rich elements in multiple "soft" nuclear explosions and in pulsed reactors has been studied [14]. We have found that in the first case the both gaps may be easily bypassed and, thus, a measurable amount of the neutron rich long-lived superheavy nuclei may be synthesized. For the second case we have formulated requirements for the pulsed reactors of the next generation which could be also used in future for the production of long-lived superheavy nuclei.

In Fig. 4 the experimental data and calculated yields of transuranium nuclei are shown for the test thermonuclear explosion "Mike" [13] assuming 1 $\mu$s neutron exposure of $1.3 \times 10^{24}$ neutrons/cm$^2$ with subsequent one-month decay time. Note that elements 99 and 100 (Einsteinium and Fermium) were first discovered just in debris of the "Mike" explosion. In this case the Fermium gap does not influence the yields of nuclei with $Z > 100$.

The possibility of generating two or several nuclear explosions in close proximity of each other to increase the resulting mass number of the synthesized nuclei could be also considered. Such a possibility is illustrated in the upper part of Fig. 5. In the bottom part of this figure the probabilities of heavy element formation are shown for one, three and ten subsequent short-time (1 $\mu$s) neutron exposures of $10^{24}$ n/cm$^2$ each following one after another with

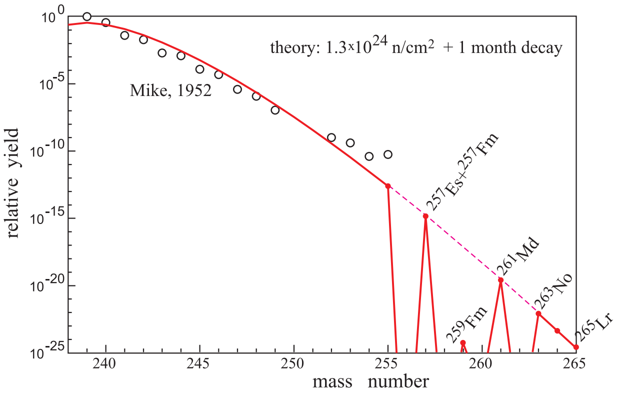

**Fig. 4.** Experimental and calculated relative yields of heavy nuclei in the test nuclear explosion "Mike"[13].

time interval of 10 seconds with final one month waiting (needed to perform some experimental measurements). These results demonstrate that multiple rather "soft" nuclear explosions could be really used for the production of noticeable (macroscopic) amount of neutron rich long-lived superheavy nuclei. Leaving aside any discussions on possibilities of such processes and associated technical problems, we want to emphasize a sharp increase of the probability for formation of heavy elements with $Z \geq 110$ in the multiple neutron irradiations (enhancement by *several tens* of orders of magnitude). This probability is high enough for some superheavy elements (see the region above the dotted line in Fig. 5) to perform their experimental identification.

We studied the same process of multiple neutron exposures realized in pulsed nuclear reactors. The pulse duration here could be much longer than in nuclear explosions (up to few milliseconds). In spite of that, the neutron fluence usually does not exceed $10^{16}$ n/cm$^2$ in existing nuclear reactors ($10^{19}$ n/cm$^2$s during one millisecond pulse). Thus, the multi–pulse irradiation here corresponds, in fact, to the "slow" neutron capture process, in which new elements with larger charge numbers are situated close to the line of stability and finally reach the Fermium gap where the process stops.

The situation may change if one would be able to increase somehow the intensity of the pulsed reactor. The neutron fluence in one pulse and frequency of pulses should be high enough to bypass the both gaps of short-lived nuclei on the way to the island of stability. The specification of the high–intensity pulsed reactors of next generation depends strongly on properties of heavy neutron rich nuclei located to the right of these gaps. These nuclei are not discovered yet, and undoubtedly certain experimental efforts should be made to resolve this problem. We have found that increase of the neutron fluence in the individual pulse by about three orders of magnitude as compared with existing pulsed reactors, i.e. up to $10^{20}$ netrons/cm$^2$, could be quite sufficient to bypass the both gaps [14].

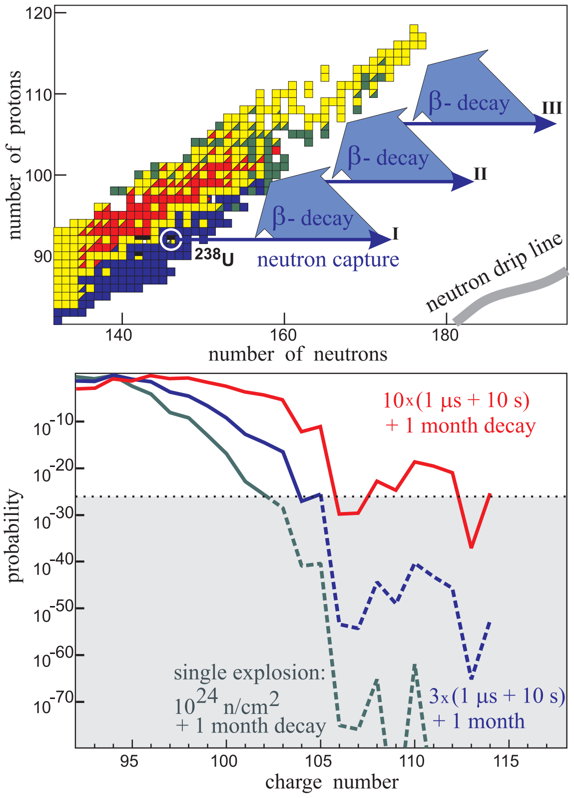

**Fig. 5.** Schematic picture for multiple neutron irradiation of initial $^{238}$U material (up) and probability for formation of heavy nuclei (bottom) in such process (one, three and ten subsequent explosions). Dotted line denotes the level of few atoms.

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